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SURFACE ACOUSTIC WAVE INVESTIGATION OF SUPERCONDUCTING FILMS,(U)

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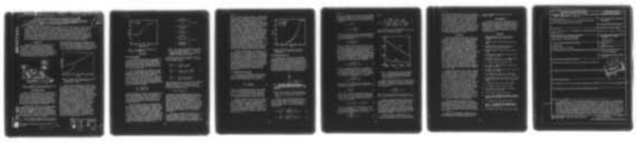

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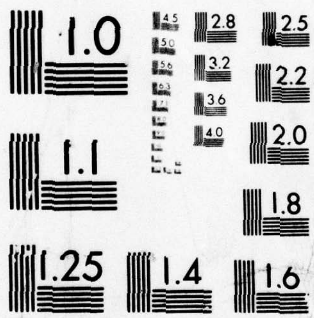

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SURFACE ACOUSTIC WAVE INVESTIGATION OF SUPERCONDUCTING FILMS

(10) Moises Levy, Harry Salvo Jr., David A. Robinson,[†] Kazumi Maki[††] and Masashi Tachiki[†††]
Department of Physics, University of Wisconsin-Milwaukee
Milwaukee, Wisconsin 53201

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ABSTRACT. The ultrasonic attenuation coefficient of 2 GHz surface waves in a 300 Å aluminum film has been measured in the normal and superconducting state. A zero temperature energy gap of 3.6 k_BT_c is found for the Al film. Microwave electromagnetic attenuation measurements in the film indicate critical fluctuation effects near T_c. However, the evidence for the contribution of fluctuation effects to the ultrasonic attenuation data of this short mean free path Al film is not conclusive. The mean free path determined from the electron phonon interaction term obtained from the ultrasonic measurement appears to be at least an order of magnitude larger than the mean free path obtained from a measurement of the electrical conductivity of this thin granular aluminum film. This large difference is believed to indicate that the mean free path determined ultrasonically is for the Al metal while the one determined electrically is for the Al-Al_2O_3 matrix structure.

Introduction

The ultrasonic attenuation coefficient due to electron-phonon interaction has been measured in a 300 Å granular aluminum film using 2 GHz acoustic surface waves. These measurements have provided information about the superconducting energy gap in the aluminum film, the electron mean free path in the aluminum film and the contribution of fluctuations to electron phonon interaction near the superconducting transition temperature T_c of the film.[1,2]

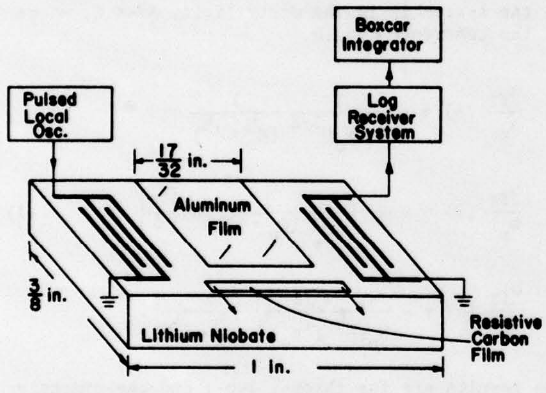

Figure 1. Schematic of system used for producing, propagating and detecting surface waves.

Experimental Results

Figure 1 shows a schematic of the experimental arrangement. An aluminum film of about 300 Å thickness was evaporated in the presence of oxygen from an aluminum wetted tungsten filament onto the lithium niobate substrate maintained at room temperature. This resulted in a granular aluminum film, composed of metal islands 50 Å in diameter embedded in an Al_2O_3 matrix,[3] with an enhanced transition temperature and presumably enhanced fluctuation effects.[4,5]

Acoustic surface waves on Y-cut lithium niobate, propagating along the Z direction were generated and detected using interdigital transducers having a fundamental frequency of 700 MHz. The third harmonic of one set of transducers, 2 GHz, was excited using a pulsed transmitter. The generated surface waves were then detected on the other set of transducers using an IF log amplifier and a Boxcar Integrator.

Since the transition temperature of the film was 1.68 ± .03 K, temperatures below 1 K were required in order to measure the temperature dependence of the attenuation coefficient through a significant temperature range. These temperatures were obtained by using a ^{3}He evaporation refrigerator that could attain 0.4 K. A carbon film thermometer was located directly on the surface of the lithium niobate substrate.

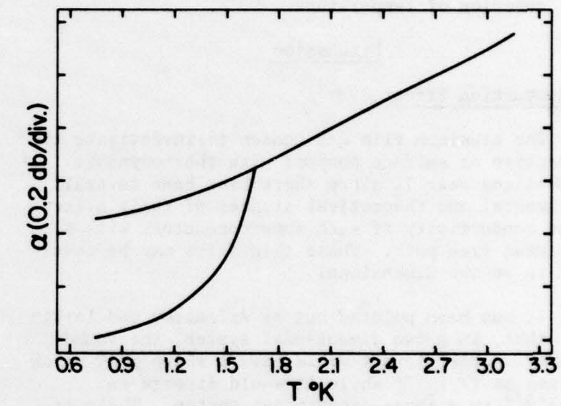

Figure 2. Surface wave attenuation as a function of temperature. Upper curve obtained in normal state of Al film.

Figure 2 shows a superposition of the attenuation in the normal and superconducting states of the Al film down to 0.74 K. The normal state was obtained by applying a magnetic field. Both superconducting and permalloy shieldings were used to reduce the earth's magnetic field below fifth milligauss when the measurements were performed in the superconducting state of Al. The d.c. resistance of this film in the normal state was 100 ohms per square. Figure 2 was obtained by locating the boxcar integrator gate on the transmitted ultrasonic pulse. By locating the gate on the breakthrough pulse, Figure 3, it is possible to observe the attenuation of the electromagnetic pulse by the aluminum film as a function of temperature. These breakthrough data were obtained at 1.6 GHz. The sharp local minimum in the attenuation is a reproducible and reversible effect occurring within experimental accuracy at the T_c measured ultrasonically. Note the rounding of the attenuation curve above T_c, indicating that critical fluctuation effects are observable and that their effect extends over a temperature range of about 0.1 K above and below T_c.[6]

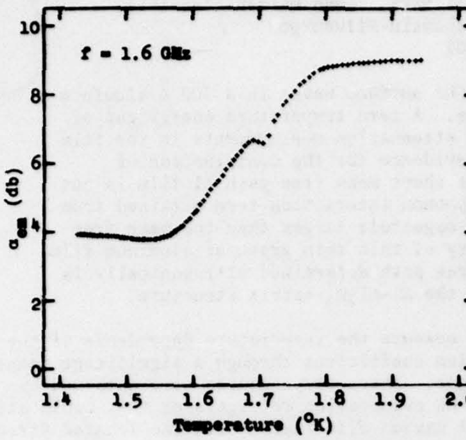

Figure 3. Electromagnetic breakthrough as a function of temperature.

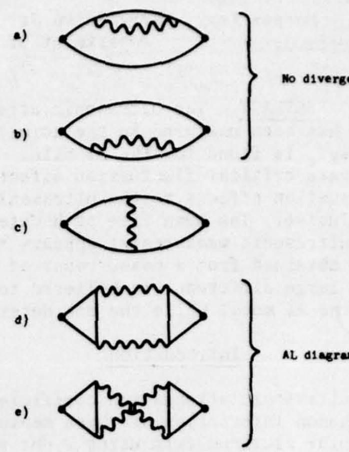

Figure 4. The lowest order diagrams that contribute to the sound attenuation. (a) and (b) do not produce any divergence at T_c. (c) is the anomalous diagram. (d) and (e) are the diagrams due to Aslamazov and Larkin.

Discussion

A. Fluctuation Effect

The aluminum film was chosen to investigate the interaction of surface phonons with thermodynamic fluctuations near T_c since there have been several experimental and theoretical studies of their effect on the conductivity of such superconductors with a short mean free path. These thin films may be considered to be two dimensional.

It has been pointed out by Aslamazov and Larkin (AL)[7] that, in a two dimensional system, the sound attenuation coefficient *would have a sharp peak which* diverges as $(T-T_c)^{-2}$ while it would diverge as $(T-T_c)^{-3/2}$ in a three dimensional system. Their expression applies only in the limit $q\ell \gg 1$, where q is the sound wave vector and ℓ is the electron mean free path. In addition, in the dirty limit, *their model* would give an effect that would be considerably smaller than the normalized fluctuation contribution to the electrical conductivity, namely

$$\frac{\alpha_{AL}}{\alpha_n} \simeq \left(\frac{k_B T_c}{E_F}\right)^2 \frac{\sigma_{AL}}{\sigma_n}$$

where the subscripts AL refers to the Aslamazov Larkin fluctuation contribution to either the ultrasonic attenuation coefficient α or the electrical conductivity σ, and E_F is the Fermi energy. Since the numerical factor $(k_BT_c/E_F)^2$ is of the order of 10^{-8} an experimental observation of this effect appears extremely difficult.

Figure 4 shows the lowest order diagrams that contribute to the sound attenuation. The wavy lines show the superconducting fluctuation propagator; the solid lines, the electron propagator. The AL attenuation coefficient is concerned with diagrams (d) and (e). In these the fluctuations carry the superconducting current; each of the two vertices accounts for a factor (k_BT_c/E_F). Diagrams (a) and (b) produce no divergence. Diagram (c) is the anomalous diagram due to Maki.[8] This diagram has no vortex of the form seen in diagrams (d) and (e) and therefore the factor of (k_BT_c/E_F) does not appear in the final result. Using diagram (c) the fluctuation contribution to the attenuation coefficient may be obtained, assuming that the system is in the *dirty limit*, $\ell \ll \xi$, where ξ is the coherence length.

$$\frac{\alpha_{fl}}{\alpha_n}(3) = -\frac{T}{2\pi N_o D^{3/2}} \frac{1}{(\epsilon_o^{1/2} + \delta^{1/2})}$$

$$\frac{\alpha_{fl}}{\alpha_n}(2) = -\frac{T}{2\pi N_o Dd} \frac{1}{(\epsilon_o - \delta)} \ln\left(\frac{\epsilon_o}{\delta}\right) \qquad (1)$$

$$\frac{\alpha_{fl}}{\alpha_n}(1) = -\frac{T}{N_o s D^{1/2}} \frac{1}{(\delta\epsilon_o)^{1/2}} \frac{1}{(\epsilon_o^{1/2} + \delta^{1/2})} .$$

These results are for three-, two-, and one-dimensional systems, respectively. In equation (1), D is the diffusion constant, N_o in the electron density of states at the Fermi level, d is the film thickness, s is the cross-sectional area and

$$\epsilon_o(T) = (8/\pi)(T-T_c) .$$

In the above, the pair breaking energy δ has been introduced following Thompson[9] to suppress the divergence in the two- and one-dimensional systems. The above contribution of the anomalous term to the attenuation is the same as its contribution to the electrical conductivity but of opposite sign. This implies that the additional contribution to the ultrasonic attenuation due to fluctuations is exactly the same as the anomalous contribution to the electrical conductivity. However, for the electrical conductivity, the Aslamazov-Largin terms also contribute significantly while they do not to the ultrasonic attenuation.

According to Aslamazov and Larkin, their mechanism would give a peak in the attenuation at the transition temperature while the Maki term would round the attenuation curve at T_c. A peak was not observed in any of the experimental measurements. In the preliminary measurements a slight rounding of the

curve near T_c was observed. However, the measurements shown in Figure 2 were obtained about a year after the aluminum film was evaporated, thus allowing the film to absorb even more oxygen and making the film dirtier. Therefore, it is possible that the anomalous contribution is completely suppressed by the dirtiness of the sample. An inspection of equation (1) with its pair breaking term, indicates that the anomalous contribution should be observed in a pure freshly evaporated sample. On the other hand, the Aslamazov-Larkin term exists independent of the electronic mean free path. Thus this accounts for the observation of the critical fluctuation in the electrical conductivity in Figure 3 while they do not appear in Figure 2. Therefore the ultrasonic attenuation measurements together with the electromagnetic fluctuation effect appear to indicate that the fluctuations of the superconducting order parameter associated with the Aslamazov-Larkin process have, in fact, an effect that is orders of magnitude smaller on the ultrasonic attenuation than on the electrical conductivity. This may be because electrical currents seek superconducting paths produced by fluctuations while the ultrasonic waves do not have this same freedom of motion.

The peak in attenuation at T_c predicted by AL is the usual result that is expected at a second order phase transition. It is produced by a relaxation process wherein, in this instance, energy is transferred to the superconducting fluctuations and is returned to the sound wave. At a temperature such that the lifetime of the fluctuations gives $\omega\tau = 1$, a peak in the attenuation coefficient would be expected. The Maki process appears to be just the normal contribution of the superconducting fluctuations to the attenuation. Namely, any superconducting portion of the film will decrease the attenuation since these reduce the excitations present which may interact with the surface waves.

B. Superconducting Energy Gap

According to the BCS[10] theory the ratio of the ultrasonic attenuation coefficient of longitudinal waves in the superconducting state to that in the normal state in the regime $q\ell >> 1$ is given by

$$\frac{\alpha_s}{\alpha_n} = \frac{2}{(e^{\Delta/k_BT} + 1)} \tag{2}$$

where q is the temperature dependent order parameter. It has been shown[11] that this relation also holds for $q\ell << 1$ for both longitudinal and transverse waves. Since a surface wave has both longitudinal and shear components and since in our case $q\ell << 1$ it is expected that relation (2) should also hold.

Figure 5 shows the normalized acoustic surface wave attenuation taken at 2 GHz. The solid line is a plot of equation (2) with a transition temperature of 1.65 K and a zero temperature energy gap of $2\Delta(0) = 3.60\ k_BT_c$. The data are fitted quite well by the BCS theory. Close inspection of the data very near T_c indicates that there might be a slight precursor or rounding just above T_c. Further experiments are in progress to investigate freshly evaporated Al films.

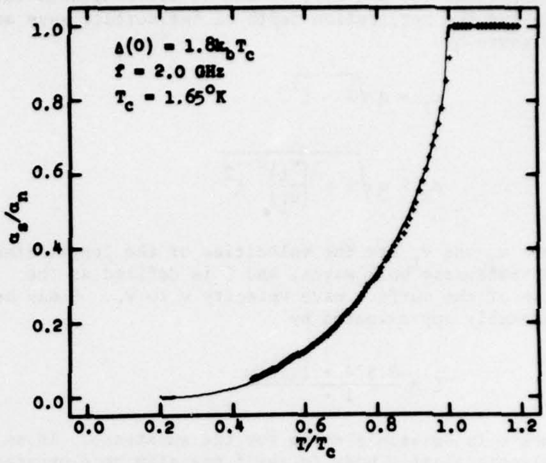

Figure 5. Normalized ultrasonic attenuation coefficient as a function of the reduced temperature.

C. Electron Mean Free Path

As may be seen in Figure 2, the attenuation change experienced by the surface wave as the film went from the normal to the superconducting state was 0.45 dB which yields an attenuation coefficient of 0.33 dB/cm. This attenuation is being produced only by electron-phonon interaction in the Al film since when the Al film becomes superconducting this part of the attenuation is eliminated. Now we have to determine the attenuation coefficient of the Al film. This may be done if we look at the following model.

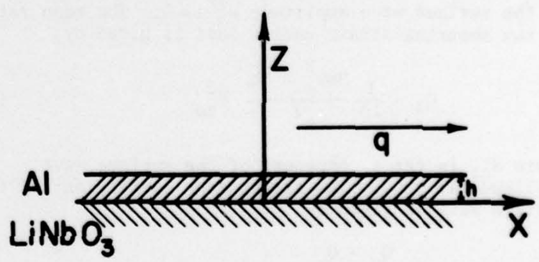

Figure 6. Model used for calculating the attenuation in the Al film. h is the thickness of the Al film. The propagation vector $\vec{q}$ is along x.

We shall use the coordinate system shown in Figure 6 where surface waves propagate along the x-axis. We shall assume that the substrate is an isotropic elastic body. The x- and z-components of the displacement vector of the surface waves are expressed as[12]

$$S_x = A\kappa_1\left[e^{\kappa_1 z} - \frac{2q^2 e^{\kappa_2 z}}{q^2+\kappa_1^2}\right]\cos(qx-\omega t) \tag{3}$$

$$S_z = Aq\left[e^{\kappa_1 z} - \frac{2\kappa_1\kappa_2}{q^2+\kappa_1^2}\, e^{\kappa_2 z}\right]\sin(qx-\omega t) \tag{4}$$

where A is a constant, and q and ω are the wave number and angular frequency of the surface wave respectively. In equations (3) and (4), κ_1 and κ_2 are constants indicating the penetration depth of the surface wave and are given by

$$\kappa_1 = q\sqrt{1 - \xi^2}$$

$$\kappa_2 = q\sqrt{1 - \left(\frac{v_t}{v_l}\right)^2 \xi^2}$$

where v_l and v_t are the velocities of the longitudinal and transverse bulk waves, and ξ is defined as the ratio of the surface wave velocity v to v_t. ξ may be reasonably approximated by

$$\xi = \frac{0.874 + 1.12\,\nu}{1 + \nu}$$

where ν is Poisson's ratio for the substrate. In an isotropic elastic body $(v_t/v_l)^2$ may also be expressed as a function of Poisson's ratio as $(1-2\nu)/2(1-\nu)$. The x and z components of the surface vibration induce compressive and shearing strains in the Al film. These strains cause energy dissipation of the surface wave to the conduction electron system in the Al film due to Pippard's mechanism.[13] When the electron mean free path is much shorter than the wavelength of the surface wave, the mean rate of the compressive strain energy loss per unit surface area is given by

$$Q_x = \frac{2}{15}\frac{Nmv_o^2\,\omega^4\tau h}{v^2}\,S_{xo}^2 \qquad (5)$$

where h is the film thickness, v_o is the Fermi velocity of the conduction electrons, N the number of electrons per unit volume, τ the relaxation time of the conduction electrons, and S_{xo} is the x component of the surface wave amplitude at $z = 0$. The mean rate of the shearing strain energy loss is given by

$$Q_z = \frac{1}{10}\frac{Nmv_o^2\,\omega^4\tau h}{v^2}\,S_{zo}^2 \qquad (6)$$

where S_{zo} is the z component of the surface wave amplitude at $z = 0$. The attenuation coefficient of the surface wave is given by

$$\alpha = \frac{Q_x + Q_z}{vE} \qquad (7)$$

where E is the energy of the surface wave per unit surface area. It can be shown that the mean kinetic energy of a surface wave is equal to its potential energy. Thus, the total surface energy is given by twice the kinetic energy

$$E = \int_{-\infty}^{0} \rho[\langle(\dot{S}_x)^2\rangle + \langle(\dot{S}_z)^2\rangle]dz \qquad (8)$$

where ρ is the density of the substrate and $\langle A\rangle$ denotes the time average of A. The value of E is calculated using equations (3), (4), and (8). Inserting the energy E and equations (5) and (6) into equation (7), we obtain

$$\alpha = \frac{Nm\nu_o^2\omega^2\tau}{\rho v^3}\,qh\,F(\nu) \qquad (9)$$

where

$$F(\nu) = \frac{\frac{1}{5}\left(1 - \frac{2ab}{1+a^2}\right)^2 + \frac{4}{15}a^2\left(1 - \frac{2}{1+a^2}\right)^2}{\frac{a}{2} - \frac{4a}{(1+a^2)} + \frac{2a^2}{(1+a^2)^2 b} + \frac{1}{2a} + \frac{2a^2 b}{(1+a^2)^2}} \qquad (10)$$

with $a = \sqrt{1 - \xi^2}$ and $b = \sqrt{1 - (v_t/v_l)^2\xi^2}$. The calculated values of $F(\nu)$ are shown in Figure 7.

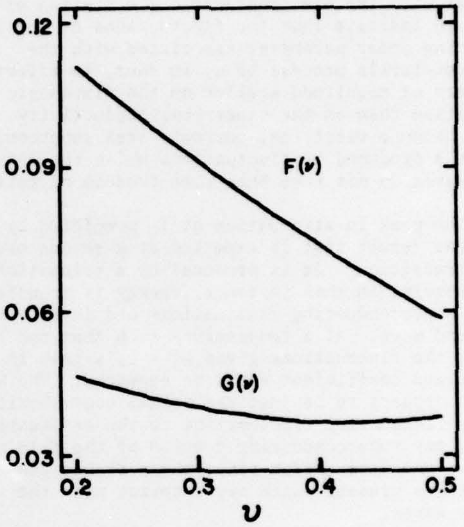

Figure 7. Graph of $F(\nu)$ and $G(\nu)$ as a function of ν. $F(\nu)$ assumes continuous displacement across the film substrate interface, equation (9), while $G(\nu)$ assumes a continuous strain, equation (11).

These results were obtained by assuming that the displacement is *continuous* at the interface between the film and the substrate. However, if continuity of the strain is assumed at the interface then equation (9) is modified to

$$\alpha = \frac{Nm\nu_o^2\omega^2\tau}{\rho v^3}\,qh\,G(\nu) \qquad (11)$$

where the only change has been to replace $F(\nu)$ by $G(\nu)$. This latter quantity is also plotted in Figure 7.

Using the measured attenuation coefficient and equation (9), we obtain the electron mean free path as 11.5 Å. With equation (11), we obtain 26.2 Å. Here we have used $N = 1.806 \times 10^{23}\ \mathrm{cm^{-3}}$, m = free electron mass, $v_o = 2.02 \times 10^8$ cm/sec, $\rho = 4.7$, $v = 3.226 \times 10^5$ cm/sec and $\nu = 0.309$.[14] The latter mean free path is consistent with a mean free path which is given by diffuse scattering over the surface of a sphere of diameter d = 50Å, in which case the average mean free path $\ell = (2/3)d = 33$ Å.

From the electrical measurements we find $\ell = v_o\tau = 1.3$ Å, using $\sigma = Ne^2\tau/m$. We would like to call this mean free path an effective mean free path

since the actual mean free path in the Al is probably 26 Å. However, this electrical mean free path reflects the fact that the electrons are tunneling from one metal island to another. Thus, the number obtained for the mean free path is really a conversion of the tunneling probability to a mean free path and the equation for the conductivity is our means of converting the measured electrical resistivity, which is limited by tunneling, into an effective mean free path. As is seen, the two values differ by a factor of 20. This appears to be consistent with a model wherein the ultrasonic surface waves sample the aluminum islands and Al_2O_3 matrix in parallel while the electrical current goes through them in series.

Conclusion

We have shown that by using 2 GHz surface waves the change in the ultrasonic attenuation coefficient produced by a 300 Å Al film as it undergoes a phase transition from the normal to the superconducting state may be measured. Thus we are measuring the contribution of the electron-phonon interaction term to the ultrasonic attenuation in the aluminum film. These measurements can be used directly to find the zero temperature superconducting energy gap of the aluminum film $2\Delta(0) = 3.6\ k_BT_c$. The data were fit quite well by the BCS expression. This is a surprising result in view of the fact that the Al film is composed of an aggregate of metal islands in an Al_2O_3 matrix. However, it fits in quite well with our result obtained from the mean free path determination which states that the ultrasonic waves sample the metal islands in parallel. Thus the Al_2O_3 matrix contributes very little to the change in attenuation and therefore the total change is just due to the metal islands. Since the mean free path in these is small, anisotropies are averaged out and according to the Anderson[15] model one should obtain good agreement with the BCS theory.

Although the raw data shows no obvious evidence of fluctuation contribution to the ultrasonic attenuation coefficient near T_c, a careful scrutiny of the analyzed data of Figure 4 near T_c shows an indication of a precursor attenuation decrease. In this graph both the energy gap and T_c were varied to obtain the best fit to the data. In all cases it was always necessary to choose a T_c which was slightly lower ($\sim$0.01K) than the temperature at which the attenuation started to drop from its normal state value. This, however, is not considered conclusive proof of the existence of a contribution of the anomalous fluctuation term to the ultrasonic attenuation coefficient. The present composition of the film was chosen, because it was known that this would enhance the fluctuation contribution to the electrical conductivity. After the theory was developed it was realized that in order to enhance the contribution of the anomalous term to the attenuation coefficient a pure Al film would be preferable. Experiments are in progress in order to attempt those measurements.

The electron mean free path determination, demonstrates that these measurements of the electron-phonon interaction term in thin metallic films yield information about the films which is complementary to that obtained by other means. The electrical conductivity yields a series averaged mean free path while the ultrasonic measurement yields a parallel averaged mean free path.

Finally, these measurements demonstrate that highly polycrystalline samples of materials that may only be prepared in thin film form may be successfully investigated using surface waves in the gigahertz range.

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† Permanent address, Texas Instruments, Dallas, Texas.

†† Permanent address, University of Southern California, Los Angeles, California.

††† Permanent address, Tohoku University, Sendai, Japan.

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20. ABSTRACT

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